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LONG-TERM EFFECTS OF AIR POLLUTION—

A SURVEY

June 1970
CEM 4029-400

G. D. Robinson
Principal Investigator

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PREFACE

This report stems from the desire of the National Air Pollution Control Administration to assess the need for research into the long-term geophysical and biological effects of air pollution. It takes the form of a survey of the problem areas and a broad delineation of the lines on which useful research could be pursued.

The report was prepared within The Travelers Research Corporation by Dr. G. D. Robinson with assistance from Miss Marcella Czarnecki and Mr. Marshall A. Atwater. Mr. Atwater's substantial contributions to Section 5 include original work on the radiative effects of aerosol, part of an investigation which I hope will be published shortly. The subject area of precipitation physics was dealt with, under a sub-contract, by a team at Meteorology Research, Inc., Altadena, California, headed by Dr. Theodore B. Smith. Their report has been condensed and paraphrased in the main body of this document, which will have a wide distribution. Their full report is an appendix to the document submitted to the sponsor. Dr. William H. Smith, of the School of Forestry, Yale University, acted as consultant on biological matters. His advice is quoted and paraphrased in the appropriate sections and his reports are attached as appendices for the sponsor.

Many other colleagues have been questioned and consulted informally during preparation of the report, and some of them may recognize their unacknowledged contributions (though appropriate reference has been made when original scientific work is concerned).

The authors and sponsor hope that readers of this document will not hesitate to offer critical comment.

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1.0 SCOPE OF THE STUDY

We cannot begin the task of formulating a research program into the long-term geophysical and biological effects of air pollution with a clear and generally acceptable specification of what kind of effect we are concerned with. There might be less need for a research program if we could. The subject matter of such a program can perhaps better be indicated at the present time by example than by an attempt at formal definition.

1.1 Geophysical Effects

If an effect stemming from localized pollution emissions is global in nature, we are clearly concerned with it. The obvious example is the global climatic (geophysical) change which might follow an increase in the atmospheric CO_2 content. But there are other more direct and less widespread climatic or geophysical effects of pollution which are long-term. Examples are the reduction of sunlight in major cities and increase of rainfall downwind of certain industrial complexes. These "regional" and "localized" long-term effects are included in the scope of the study, both because of their immediate impact on public welfare and because we cannot, in the present state of knowledge, be quite sure that some of them do not have a reaction on global conditions.

1.2 Biological Effects

The effects of air pollutants on plants and animals may be classified into three rather broad categories. The first group contains those effects which result from relatively short-term exposures to high levels of pollutants. The consequences of these exposures are commonly termed acute effects. The second group encompasses those effects which occur as a result of exposure to comparatively low levels of pollutants over more extended time periods. These phenomena are typically labelled chronic effects. The third category of effect includes those abnormal ecological and physiological alterations in living systems which may accrue after long-term coexistence of ecosystems and air contaminants. For lack of a better term, we might classify these latter abnormalities long-term effects. Long-term effects differ from chronic effects in several regards. In the case of chronic effects, the time horizon may be measured in years whereas the long-term effects may have a time horizon of decades, centuries or greater. In the instance of gaseous air pollutants, chronic effects are presumably caused by the gas or gases acting directly on the plant or animal. With long-term effects, however, the significant agent may be a by-product of the gas, rather than the gas itself (e.g., nitrate in the case of oxides of nitrogen) and it may influence the plant or animal indirectly rather than directly. Finally, and perhaps more importantly, our knowledge concerning the ramifications of the influences of air pollutants are greatest in the instance of accute effects, much less in the case of chronic effects, and almost nil in regard to long-term effects.

1.3 Areas of Research

The study of air pollution involves study of the emission of pollutants, their transport and transformation in the atmosphere, their deposition on land and ocean, their interaction with the biosphere, and the ways in which they modify the physics

and chemistry of the unpolluted atmosphere. All these study areas are involved when we investigate long-term effects. We are also concerned with monitoring, which is taken to mean the purposeful, controlled, continuing observation on a global or local scale of a pollutant, or of an established or suspected effect of a pollutant.

It is convenient at this point to stress the need for continuing study of likely future emissions, in the light of population trends, changing patterns of life, economic and natural resources (for example, availability of fossil fuel), available technology, and available instruments for abatement on an international scale. Actual and projected emission data will at some point be needed as an input into quantitative research on long-term effects. Equally, since there is a possibility (and it cannot be described at this time, as more than a possibility) that physical, chemical, mathematical or biological studies might at some stage indicate disastrous consequences if emission of some pollutant were to continue, these studies should be paralleled by technological-economic studies on if and how the world's increasing energy conversion needs could be met with varying constraints on pollution emissions. No further reference will be made to this subject.

2.0 THE MAJOR POLLUTANTS: THEIR EMISSION AND ROLE IN THE ATMOSPHERE

A recent report by E. Robinson and E. C. Robbins [1] gives an excellent summary of the incidence, chemistry and physics of the major pollutant gases. This is the main source of the following brief summary of some of the facts which seem particularly relevant in planning an investigation of long-term effects.

2.1 Carbon Dioxide

This is not the place to discuss in depth the literature on the carbon dioxide content of the atmosphere. It has rather cursory treatment in Robinson and Robbin's report. The comprehensive work of Bolin and Keeling [2] has been updated in a publication by Bolin and Bischof which became available after completion of the text of this report. The Bolin and Keeling paper, with others which are referenced in it, covers the major factors of interest without perhaps doing full justice to the role of the biosphere in the CO_2 turnover. The broad facts which appear to be established are that there is an increasing annual production of CO_2 from fossil fuel combustion of about 1×10^{10} tons per year at present, and that there has been for many years a steady increase in the atmospheric CO_2 concentration which in the mid-1960's was around 7×10^{-7} per year, accounting for about half the annual production of CO_2 by combustion. There is recent evidence (Machta, personal communication) that this factor may have fallen to about one-third the annual production. There is an exchange of CO_2 between atmosphere and biosphere—continental and oceanic—involving a quantity of CO_2 greater than that produced by fossil fuel combustion. There is a very large oceanic reservoir of dissolved CO_2 and CO_2 "fixed" as carbonate, with a chemistry, involving silicates and phosphates as well as carbonates, which has been studied in some detail but which is not yet able to account quantitatively for the ocean-atmosphere exchange of CO_2 and its geographical variation or for the fate of that fraction of the CO_2 produced by combustion which does not appear to be retained in the atmosphere.

Estimates of the quantity of CO_2 fixed in the biosphere are about 25 percent of the total atmospheric content. There is a detectable seasonal effect of the biosphere on the atmospheric content, but nothing is known about secular change.

A recently reported observation (Seiler and Junge [3], quoting Georgii) introduces a complication, and if confirmed, may require a re-examination of the mechanism of the atmospheric transport of the CO_2 produced or exchanged at the surface. This is an apparent change in the CO_2 content of about 0.2 percent, "a small but distinct difference," as the tropopause is crossed—the stratospheric content being lower. Apart from this, the relatively small systematic temporal and spatial variations in CO_2 content are so satisfactorily explained by Bolin and Keeling's simple diffusive transport model that we might expect a comprehensive general circulation model to account for them quantitatively.

2.2 Particulate Content of the Atmosphere

There are recent summaries and bibliographies by Horak [4] Shah [5] and Twomey and Wojciechowski [6] dealing with the particulate content of the atmosphere.

A few notes concerning the role of pollutant aerosol are added here. Radiative effects are discussed in Section 5 and effects on precipitation processes in Section 6.

We cannot entirely separate "aerosol pollution" from "natural aerosol" even by definition—the New Zealand meteorologist who remarked that he had never seen clean air north of 40°S did not necessarily mean unpolluted air. Avoiding pedantic classification, we can recognize three broad types of natural aerosol—volcanic dust, wind raised dust and water-soluble nuclei. The first two categories are clearly enough definable but soluble nuclei are produced by all combustion processes, as well as by seaspray and by chemical reactions in an "unpolluted" atmosphere, and the extent to which any population is "natural" must always be in some doubt.

There is evidence of a fairly homogeneous population of soluble nuclei in air near the surface in the world's remaining empty spaces, and growing evidence of a similar state of affairs in the stratosphere. Some of the evidence is indirect—it comprises the measurements of atmospheric electrical conductivity and the earth's potential gradient which were commonly made on scientific expeditions in the early years of the century. Conductivity decreases, and the potential gradient increases, with the nucleus and particle content of the air because the particles capture small ions and reduce ionic mobility. Atmospheric potential gradient, indicating the concentration of condensation nuclei, is an excellent man-detector or rather combustion-detector. (Radioactive fallout has confused this issue since World War II.) There are some more direct observations of particulate content of air over the oceans but realization of the importance of size distribution and the practicability of determining it readily are developments of the last 20 years, and we may in time find the electrical observations made by early Antarctic explorers and the cruises of the "Carnegie" a useful indication of the particulate content of air in the earliest days of the population explosion. There is evidence in recent NCAR observations in Panama and over the adjacent sea that the smaller "natural" soluble nuclei, Aitken nuclei of radius <0.1 μm, are largely $(NH_4)_2\ SO_4$ particles formed by oxidation of H_2S or SO_2 in the presence of an excess (around 10 ppb) of free NH_3.

The larger soluble nuclei act as cloud condensation nuclei (Section 6). They appear to have a "natural" concentration over the oceans around 100 cm^{-3}, but their number is much more variable in populated continental regions—from several hundreds to several thousands per cm^3. Nevertheless, according to Squires [7], man's contribution to the number of cloud condensation nuclei is only a few percent.

It is very difficult to estimate the life-time of soluble particles in the atmosphere. Twomey [8] has estimated that air masses moving from continent to ocean achieve the typical maritime concentration in about three days. A similar time seems to be available for the processes, whatever they may be, which produce the extreme clarity of Arctic air. There is no entirely satisfying explanation of this cleansing process.

Wind-raised and wind-blown dust, the other particulate component of the troposphere not the result of industrial processes, may be natural or artificial, in the sense of resulting from cultivation. Wind-blown material from desert land and semi-arid or temporarily arid cultivation seems to be more pervasive than was once suspected: indeed it is suggested in Appendix 1 (on the basis of an unpublished survey) that only a few percent of the total mass of atmospheric particulate is man-made. It

is often detected by its effects on solar radiation and it appears to be a major source of ice nuclei. Together with stratospheric fall-out and industrial particulates, it is to some extent preserved in the annual layers of permanent snowfields so that there is a possibility of investigating its secular changes in the past. Russian workers claim to have traced the industrial development of the USSR in the snows of the Caucasus.

A more recent development, potentially of considerable biological importance, is the long-distance transport of persistent insecticides, either on raised dust or directly on the base used in crop dusting operations. Much pollutant insecticide is water-borne but it seems that a proportion, not negligible in some localities, has been an air pollutant at one stage of its unfortunately long life.

The modification of the earth's albedo by volcanic dust is one of the mechanisms postulated to cause climatic change. A recent incident—the stratospheric dust cloud following an eruption on Bali in 1963—has been well documented and is the first such occurrence which has been extensively investigated quantitatively. Dust content of the middle stratosphere appears to have temporarily increased by an order of magnitude, with a half-life of a year or two. The extent to which the sulphate aerosol discovered in the stratosphere by Junge is of volcanic origin is not clear. This aerosol, which appears to have a maximum concentration in the height range 15 km to 25 km has been directly sampled and chemically analyzed and has been detected optically by several techniques—searchlights, lasers, and scattering from the solar beam at twilight. There is as yet no convincing explanation for this concentration, and although it seems unlikely, pollutant SO_2 may have some part in its formation. For this reason, it seems advisable that it should be extensively observed for a number of years to determine any secular trend.

2.3 Carbon Monoxide

There is no reason to expect CO to produce any direct geophysical changes at the volume concentration of about 10^{-7} which appears to be its present atmospheric level in regions remote from pollution sources. Furthermore, there is no evidence that this concentration is increasing. Atwater has confirmed that at this concentration possible radiative effects are far less than our uncertainty of the radiative effects of H_2O, CO_2, cloud and aerosol. CO cannot be ignored in the context of long-term changes, if only because we do not know how it is removed from the atmosphere, or whether or not there is a natural source comparable in magnitude with the very considerable pollution source. Robinson and Robbins [1] summarize the data on pollutant emission and distribution in the atmosphere—we see from these data that CO production by combustion processes, about 2×10^8 tons per year, is 1 to 2 percent of CO_2 production so that if CO is removed from the atmosphere by a process leading ultimately to its oxidation to CO_2, it will add negligibly to the CO_2 pollution load.

There is a possibility of biological effects connected with CO. It has low solubility, and there are some reports of saturated or even super-saturated ocean water and large concentrations associated with certain aquatic plants, and there has been speculation on the possibility of biological sources, as well as of biological sinks of the gas.

Recent observations (Seiler and Junge [3]) and theories (Hesstvedt [9]) (Pressman and Warneck[10]) may, if confirmed, remove at least some of the uncertainties. The observations, made on commercial aircraft, indicate a sudden decrease of CO content on passing from the troposphere to the stratosphere; the theory explains this as the result of a series of photochemical reactions in which CO is involved in the O_3 - H_2O photochemistry in the lower stratosphere and ultimately oxidized to CO_2 with destruction of O_3. The theory has only been briefly reported and there are some puzzling features which may be resolved in a fuller publication. If the theory is quantitatively sound detailed observations of CO concentrations in the region of the troposphere will be of considerable meteorological interest, but not in the context of long-term geophysical change. (If CO were removed by a different photochemical oxidation in the lower ionosphere there might be cause for concern, since in this region photochemistry and ionization are interrelated.)

Whilst the fate of CO remains unexplained, it is a reasonable subject for research in the context of long-term effects of pollution, if only because of the magnitude of CO emissions and the social and economic implications of substantial control. At present it seems reasonable to wait for clarification of the suggested photochemical removal process, though this may well require future measurements in the tropopause region.

2.4 Sulphur Dioxide

There is no reason to expect geophysical influence of pollutant SO_2 in its gaseous form. It now seems well established that the volume concentration of SO_2 in the upper atmosphere and over the oceans and remote land areas is as low as 2×10^{-10} (Robinson and Robbins [1]); Pate (private communication). At this concentration its radiative influence is negligible. As with CO, interest from the point of view of long-term effects centers on the methods by which it is removed from the atmosphere, but unlike the situation in respect of CO, the difficulty is not to find plausible removal mechanisms but to decide which among many possibilities are significant. From the geophysical point of view, the interest is in the formation and persistence of sulphate aerosol; from the biological point of view, in the accumulation in soil and surface waters of sulphuric and sulphurous acids and their salts following rain and fallout.

SO_2 is a short-lived atmospheric constituent. Pollutant production of 1.5×10^6 tons per year and a concentration of 2×10^{-10} require a mean residence time (as SO_2) of about 2.5 days. Indirect estimates of the life-time of SO_2 in highly polluted regions are much shorter, ranging from Meetham's 12 hours (southern England 1950), to a half-life of 1 to 3 hours estimated for the State of Connecticut (Hilst [11]). There have been reports of half-lives as short as 20 minutes observed by European investigatore, but details are not yet generally available. There is little doubt that variations in pollutant aerosol concentration have some influence on the variation of these figures, which suggest that acidic rain-out and fallout could be a problem at distances varying from a few tens to a few hundreds of miles downwind from major pollution sources (i.e., about 24 hours travel).

Persistent geophysical effects are more likely to be associated with the more persistent sulphate aerosol than with the gas. There is some evidence that over some seas and tropical land areas, the condensation nuclei are largely $(NH_4)_2SO_4$, associated

with a free NH_3 content of about 10 ppb (Pate, personal communication), and Junge identified $(NH_4)_2SO_4$ as an important constituent of the stratospheric aerosol. It is not clear to what extent pollutant SO_2 contributes to the "background" 0.2 ppb of SO_2 in remote areas, or to the formation of the SO_4 ion in these areas and in the upper atmosphere. There is a sufficient biological source of H_2S, and plausible oxidation mechanisms, to account for current levels of the "background" sulphate aerosol without the intervention of pollutant SO_2. The residence time of sulphur in the form of sulphate aerosol is currently estimated as being of order tens of days in the troposphere, and hundreds of days in the stratosphere. A question of major geophysical importance in the very long term is whether there is a mechanism for net transport of pollutant SO_2, even in very small proportion of the total output, into the stratosphere. Alternatively, or in addition, is there a mechanism for transport of sulphate aerosol from troposphere to stratosphere in amounts even very slightly greater than the loss by fallout or mixing?

2.5 Nitrogen Oxides

N_2O is a long-lived, fully-mixed constitutent of the atmosphere. It is not significantly an industrial pollutant. NO is a combustion product, particularly of combustion at high temperatures, and is readily oxidized in the atmosphere to NO_2 by reaction with O_3—a reaction so fast that even at the low (order 10^{-8}) concentrations of O_3 in the lower atmosphere the NO_2 concentration is significant. NO_2 is radiatively important—it absorbs solar radiation in the visible (with photochemical decomposition). It may, in fact, be responsible for a considerable part of the absorption of solar radiation in urban atmospheres elsewhere attributed to aerosol (Robinson [12]). It is a key consistituent of photochemical smog. Because of its high reactivity, it does not seem to present problems on the global scale, but it could have significance in chronic and long-term biological effects in the vicinity of large cities.

Photochemical reactions involving NO are of prime importance in the very high atmosphere, and touch significantly on human activity (communications) in their control of ionization in the lower region of the ionosphere. Again, because of high reactivity, it seems unlikely that pollutant NO or NO_2 from the surface could be transported to the ionosphere.

2.6 Ozone

Ozone is a constituent of the unpolluted atmosphere, being formed and destroyed photochemically. It is a highly reactive gas, particularly liable to destruction on surfaces. It is considered that the normal tropospheric concentration, of order 10^{-8}, represents an equilibrium between transport from a predominantly stratospheric source and destruction at the surface. One very fast homogeneous reaction $O_3 + NO \rightarrow O_2 + NO_2$ may be important even in air which might otherwise be considered unpolluted.

Ozone is also a pollutant of major importance and is unusual in being a secondary pollutant formed in a photochemically initiated chain reaction—the "photochemical smog" process. In smog conditions ozone concentrations may be as high as 5×10^{-7}. Smog does not seem to be a significant factor on the global scale, but it certainly affects city climates and appears to be responsible for widespread chronic and acute

damage to plants in the areas which it affects, so that the possibility of secondary "long-term" biological effects must be considered. The associated aerosol—polymerized organic nitrates?—appears also to be removed locally, but the possibility that a very small proportion of it is long-lived and mixed on the global scale cannot be dismissed.

Speculations concerning artificial climate modification have included some on possible manipulation of the stratospheric ozone layer—"punching a hole in the ozone" is a phrase which has been used. Since the layer is a most effective u-v filter, its removal would be biologically disastrous. The proposed mechanism is by introduction of a substance very reactive with O_3 (e.g., NO). Published discussion follows the consequences of such an introduction on the stratospheric photochemistry only to the first step of the chain and it is difficult to see how anything more than a change in the distribution of the O_3 with height could result. Normal pollutant materials to be considered in this context are sulphate and other aerosol, and oxides of nitrogen, and the possibility now arises that the development of supersonic transport might introduce both NO and hydrocarbons into the lower section of the ozone layer. The first sign of a "threat" of modification of the O_3 distribution might be an increase in the stratospheric content of these materials.

2.7 Water and Heat as Air Pollutants

"Waste" heat is an inevitable consequence of the utilization of the energy stored in fossil fuel or the atom, and it dissipates in the atmosphere, surface layers of the earth, and surface waters. An estimate of the level at which it might induce global climate change is not difficult to make. The average rate of absorption of solar energy is about 250 watts m^{-2} and simple climatic models suggest that variations of 1 percent of this might have serious consequences. This is only a few watts per square meter but is the equivalent of about half a million 1000 MW generating plants distributed throughout the world and more than 100 times man's current level of energy conversion. We, therefore, need not yet consider "heat pollution" as a global problem. It is, however, already significant locally—the air temperature of large cities is notably modified, by direct heating, from that of the surrounding countryside, individual smoke-stacks frequently generate cumulus clouds, and precipitation patterns may be modified. It is conceivable that, if some current predictions of population trends are sound, "thermal air pollution" might have more than local climatic effect through modification of such significant features as the sharp temperature gradient near the eastern seaboard of North America in winter. Current atmospheric models could be used to investigate these possibilities by introduction of an artificial surface heat source. From the geophysical point of view, the exact mode of disposal of the waste heat is probably not very important, but persistent local climatic and biological effects could be quite different from dry cooling towers, wet towers, or cooling to bodies of water. "Water pollution" is already producing unpleasant modification of fog and drizzle frequencies near badly sited wet cooling towers, and "thermal pollution" of streams is leading to biological changes.

Some concern has been expressed over the introduction of H_2O into the high troposphere and lower stratosphere by aircraft. There have been suggestions of a significant increase in cirrus cloud amounts in some localities, resulting from aircraft

condensation trails and some confirmation of this has recently been found by search of standard meteorological records (Machta, private communication). It has been argued that circumstances in which condensation trials persist or grow, and in which cloud would not otherwise form, are meteorological rarities since they require high relative humidity and absence of appreciable vertical motion, either up or down.

In the lower stratosphere, where supersonic transport aircraft will operate, the existing H_2O concentration is low—2×10^{-6}. This is almost certainly a level set by atmospheric dynamics—it corresponds to saturation at the coldest tropospheric temperature. Residence time of water in the lower stratosphere is probably about one year. With this residence time computation shows that commercial operation of one large transport aircraft in the lower stratosphere would increase the water content by a factor 10^{-4}. Some traffic projections have suggested that in 20 years time there may be several hundred such aircraft in operation so that the H_2O concentration could be changed by several percent. It would, therefore, be prudent to monitor the water content of the lower stratosphere, and to investigate theoretically the radiative and other effects of a few percent increase in concentration.

3.0 CLIMATE, CLIMATIC CHANGE, AND THEORIES OF CLIMATE

Considerable publicity has been given to some predictions of major changes in world climate consequent on continuing emission of pollutants. In this section, the nature of climatic change, the possible mechanisms of change, and the mathematical models which are tools for the investigation of climate are examined briefly.

3.1 Climate and Climate Change

When we come to investigate climatic change, we are faced with an initial difficulty of definition which has far-reaching consequences. The climate of a locality may be described by statistics of certain weather elements. Let us take air temperature over central and sountern England, as an example (because observations have been made there for a long time). Craddock [13] has examined about 100 years of the instrumental record of daily mean temperature at Kew (London) and Manley's series of about 250 years of annual mean temperatures of central England, using numerical filters which isolate the variance connected with various periodicities. Eliminating diurnal and annual periodicities and their subharmonics, Fig. 1 illustrates his findings. There is significant variance at the longest period plotted. The 250-year-long series of observed temperatures is not stationary[1]. If we define climate, as it has been conventionally defined,by the mean and variance of a 30-year series of observations, then the climate of central England has been subject to continuous change for the past 250 years; if we look before the instrumental record we find clear historical and geological evidence that climate change is a global phenomenon, and was so before man appeared. Early in the present millenium a pastoral community was able to maintain itself in southern Greenland. Twenty thousand years ago ice covered much of Europe and Canada. Climatic change is obviously not necessarily dependent on man's activity. This is not to say that man cannot change, or indeed is not changing climate, but we have insufficient statistics of "natural" changes to allow us to recognize artificial changes on the global or continental scale by statistical techniques, and it is the essence of the problem that we will now never have them.

We, therefore, turn from exclusively statistical to physical methods, though if climate is statistically defined we cannot completely exclude statistical considerations. If we understand in detail the dynamical and thermodynamical equations of the atmosphere and can solve them, we can investigate the consequences of the sort of perturbation which man's activity might introduce into the initial conditions. We have made enormous progress in this direction, best exemplified in the work of Smagorinsky's laboratory (e.g., Miyakoda, et al. [15]), but there is a fundamental difficulty which has been most clearly exposed by Lorenz[2] [16]. The behavior of the atmosphere is

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[1]The continuous line in Fig. 1 is the spectrum of a stationary series with a long time scale proposed by Charnock and Robinson [14] as an empirical approximation to many meteorological time-series. It is fitted to Craddock's spectrum at a period of 30 days.

[2]I first encountered the idea many years ago in an unpublished review by R. C. Sutcliffe and, no doubt, it has been intuitively recognized by most meteorologists; but it is, surprisingly, not a prominent feature of meteorological literature.

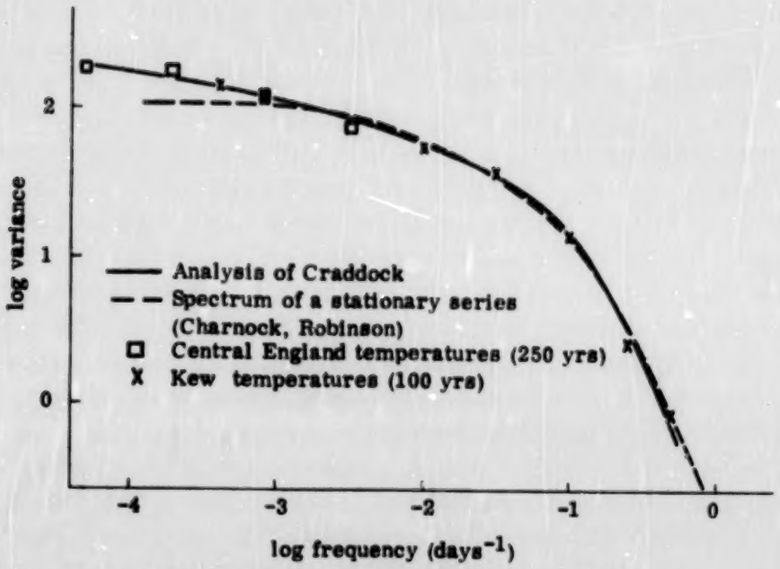


Fig. 1. Spectrum of temperature variations.

expressed as a closed set of equations. This set of equations has numerous solutions, each representing a possible state of the atmosphere. Climate can be mathematically defined as the statistics of the solutions of these equations. If a unique (stationary) set of such statistics exists, the system governed by the equations is said to be transitive. The equations governing the state of the atmosphere certainly include non-linear differential equations, and it is known that the uniqueness of long-term statistics of solutions of sets of such equations is not assured. Statistically different sets of solutions might, for example, develop from different initial conditions. If a unique set does not exist, the system is said to be intransitive. Lorenz introduces the concept of the "almost-intransitive" system, one for which an infinite set of solutions exists which is independent of the initial conditions but for which large but finite sub-sets exist which are very dependent on initial conditions. Lorenz postulates that the atmosphere may be an almost-intransitive system. The consequences for the student of climatic change are best stated in his own words—"For one thing, the mere existence of long-term climatic changes cannot by itself be taken as proof of environmental change; alternative explanations are now available. Finally, what about the not unlikely possibility that the atmosphere would be almost-intransitive if the environmental influences were constant, while at the same time external environmental changes actually are taking place? The effect of these changes will then be harder to detect, and causative connection will be more difficult to establish. For example, an environmental change which ought to bring about a 2°C temperature rise might occur just at the time when the temperature was in the process of falling 2°C as a result of almost-intransitivity. The environmental change might then go unnoticed simply because no one would see any reason to look for it."

With these considerations in mind, let us take a superficial look at climatic trends in the present century (we will need to look a little more closely later). Over that part of the world where air temperature is comprehensively observed, it is reasonably well established that temperatures increased over a 30- or 40-year

period, ending some time between 1940 and 1955. It is also reasonably well established that the CO_2 content of the atmosphere increased during the same period, by an amount not inconsistent with the rate of production by consumption of fossil fuels. The two trends were associated, and computation using the radiative transfer equations showed that quantitatively the CO_2 increase could explain the temperature rise. Many scientists did not hesitate to say that it did explain the temperature rise. A few did not hesitate to extrapolate the trend and predict a man-made climatic change which would melt the polar ice and drown many of the world's major cities. More cautious meteorologists recognized the inadvisability of the extrapolation but had to agree that the argument connecting the two trends was physically sound. But by 1960 it was becoming clear that the temperature was falling, and somewhat later it was noted that atmospheric turbidity was increasing, not only in the immediate vicinity of cities but over quite wide areas of Eurasia and North America. The increased turbidity correlated well, geographically, with the emission of pollutant aerosols, and the decreasing temperature was tentatively ascribed to an increased reflection to space of solar radiation by man-made pollution. The argument is plausible but, basically because of lack of knowledge of certain physical properties of the aerosol, it cannot be developed as precisely as was that concerning CO_2, and perhaps for this reason there has been less talk of a man-made ice-age than there was of a man-made deluge.

The relation of these recent events and the conclusions of Lorenz quoted above is obvious. Man has detectably changed the constitution of the atmosphere; in one respect globally, in another at least on a sub-continental scale. At the same time, there have been minor variations of climate. Our present knowledge of atmospheric processes suggests that these changes are what would be expected to follow from man's interference with the atmosphere. But they are also quite compatible with what we know of the statistics appropriate to an atmosphere of undisturbed constitution and they are also compatible with the possible behavior of a dynamical system as complex as the atmosphere. Climate has changed and will change, and man may never know to what extent he has contributed to or inhibited the change. Recognition of this has quite important consequences in the planning of research on the climatic effects of pollution.

3.2 Simple Climatic Change Models

There have been some attempts to estimate climatic effects of changes in the input of solar radiation (either by a change of solar constant or of global albedo) by very simple methods. They must be considered, if only because of the nature of some of the conclusions which have been drawn from them. The prototype of such models appears to be that of Sawyer [17 and 18] who treated the problem "very crudely as though the atmosphere consisted of two blocks of uniform temperature between which heat was transferred at a rate depending on the temperature difference." On this model a one percent decrease in absorbed solar radiation caused a decrease of temperature in the equatorial section of about 0.75C and in the polar section of about 0.6C. In a second application of the model, Sawyer reduced the mean annual solar input by one percent in regions poleward of 50° latitude and computed a temperature change in this sector of about 0.2C.

This model is illustrated in Fig. 2 in which have been inserted values of radiative input consistent with the most modern estimates of the solar constant, the global albedo as measured by satellites, the radiative temperatures measured by satellites, and the estimates of heat-transfer by ocean and atmosphere across latitude 50°. This can be used as an educational toy for simple numerical experiments. The basic assumption of the models under discussion is the relation between advective transport and temperature difference. The toy can be used to illustrate the difficulty of preserving a balance for arbitrary variations of the heat input given a simple, e.g., linear, relation between heat transport and temperature difference. It gives some appreciation of the potentialities and pitfalls of Sawyer's model and its elaborations.

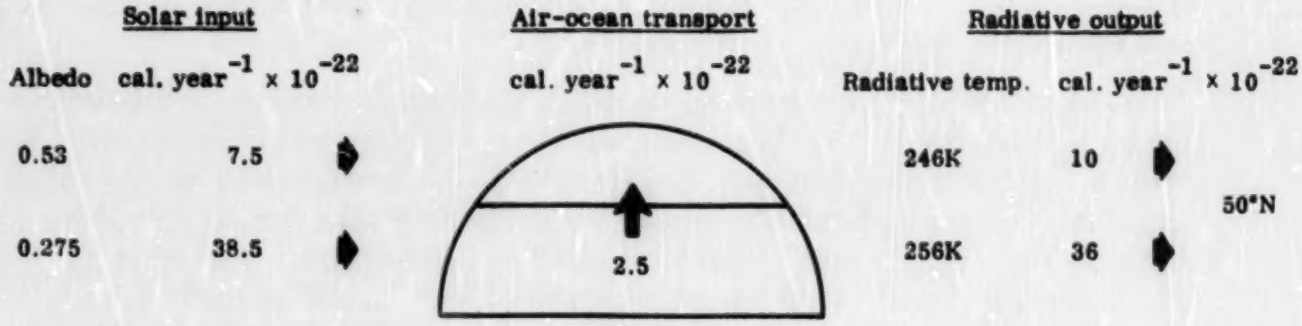


Fig. 2. Basis of the simplest climate model.

Two of the elaborations, described by Sellers [19] and by Rakipova [20]*, are directly applicable to the problem of the climatic consequence of a change of albedo. The simpler model (Sellers) carries only sea-level temperature as an (independent) indicator of climate. The type of conclusion which he draws is typified by: "If all other variables are held constant, a decrease in the solar constant by about 2 percent would be sufficient to create another ice age with the ice-caps extending equatorward to 50°." Rakipova's model is more complex in detail (though not in principle) than Sellers', and carries temperature at various heights in the atmosphere. One of her conclusions is that a one percent decrease in the solar input would result in a temperature decrease varying between 0.3C at the equator to 1.4C at the poles. The apparent difference between this and Sellers' more startling conclusion is caused by a difference in the treatment of the relation between temperature and albedo. This relation is a key factor in the models. According to Sellers, the albedo in all latitudes

*Rakipova's work exemplified the approach to this problem by the Leningrad group under M. I. Budyko.

would be expected to increase as temperature decreases. This is a destabilizing mechanism—a drop in temperature reduces the heat input. The relation proposed is empirical; the physical cause appears to be the high albedo of a snow- and ice-covered surface. The weakness of the relation is that it does not seem adequate to cover the effect of the cloud. Lower temperatures lead to a lower atmospheric water content which might mean less cloud and lower albedo. This type of control of albedo produces a stabilizing mechanism, lower temperatures leading to greater heat input, and the possibility that the planet earth is behaving as an inefficiently stirred thermostat. In Rakipova's computation, cited above, the albedo is not changed, but in the text of her paper she quotes a "warm season" global albedo of 0.410 and a "cold season" global albedo of 0.384. Both figures may be too high, but the overall relation proposed is a stabilizing relation.

The temperature-albedo relation is the sensitive problem in the Sawyer-Sellers-Rakipova type models, and there is some prospect of an empirical solution to it when many more data have been collected from meteorological satellites, but the models have also a major difficulty in principle. Rakipova states this clearly early in her text and does not allow it to inhibit her further. She states, "We are analyzing a purely zonal situation for which (the mean meridional and vertical velocities) = 0." One cannot accept this "purely zonal situation" as a reasonable model for investigation of the effects of heat transfer between latitudes. Sellers apparently shares this view but does not appear to circumvent the problem. He states "the inclusion of the mean meridional motion is necessary in order to avoid having to deal with negative diffusivities," but the mean meridional motions he postulates do not satisfy mass continuity and he does not discuss the transports of potential energy consequent on realistic mean meridional circulations.

3.3 Detailed Climatic Models

The basis of modern meteorology is mathematical simulation of the atmosphere and the earth's surface as a nearly-closed thermodynamic system, and the current trend in such simulation is toward increasingly detailed realism, at the expense of computation loads which stretch the limits of foreseen technology. This type of model was initially conceived as a tool for objective weather forecasting and is often justified economically in this context, but its value in climatology was recognized at an early stage. Reservations concerning the validity of this approach to the detailed forecasting problem (Robinson [21], Lorenz [22]), even if sound, are not related to the use of the models as generators of climatic statistics. For the present purpose, a minimal description of the methods must suffice. Several groups are engaged in the development and use of comprehensive atmospheric models. Probably the most advanced and best documented model is that of Smagorinsky's group at Princeton. The following comments, superficial in the sense that they include only aspects relevant to the present task, are based on a perhaps partial understanding of this model and of that described by Bushby and Timpson [23].

The mathematical basis is the conservation equations for momentum, energy and matter—partial differential equations, some of which are non-linear—the integral radiative transfer equation, the perfect gas equation of state and the thermodynamic equations describing the phase changes of H_2O. Mathematically, solution is only

possible by numerical finite difference methods; physically, initial and boundary conditions can only be supplied as averages in time and space. The magnitude of the computation load and the resolution of observations set lower limits to feasible and available spatial definition; when the space scale has been chosen an upper limit to the finite time step which can be used in solution of the momentum equations follows. A spatial resolution greater than about 500 km is probably insufficient in a model which is expected, for example, to indicate changes in the ice and snow covered areas of the globe. The corresponding maximum allowable time-step is of order 10 minutes. To seek to use such a model in straightforward continuous simulation to examine a period even as short as the 250 years for which we have some instrumental record, cannot at present be contemplated. Perhaps this will not still be true 50 years from now but we are concerned with the next few years. In this period the use of models to investigate climate modification will probably be by changing initial and boundary conditions—surface albedo, solar input, etc., or coefficients in the radiative transfer equations (i.e., atmospheric composition) and following the consequences to a quasi-steady state (or to breakdown of the computational scheme).

For the investigation of the effects of pollution, current comprehensive models have one very serious defect—they do not develop and transport cloud systems. Climatological averages of cloud distribution are used in application of the radiative transfer equations and the water-cycle is handled by assuming immediate precipitation of water in excess of saturation—the models simulate rain and snow but not cloud. There is no major difficulty of principle in modifying models of large-scale processes to incorporate generation, transport, and dissolution of cloud, but this would increase very considerably the complexity of a computation which is already probably the most extensive ever undertaken on a continuing basis. Lack of this modification is not of prime importance in the weather prediction application of large-scale models, but it is a most serious short-coming when they are used to investigate climatic change, because of the high sensitivity of climatic statistics to albedo, the sensitive dependence of albedo on cloud amount and type, and the critical relation of cloud amount and type to atmospheric motion and stability. We see that the major defect of the existing comprehensive models of the atmosphere, in their application to study of climate, is the same as that we noted in the simple models—inadequacy in simulation of the relation of albedo to other parameters of climate.

It is probably safe to assume that on the global and climatic scale, though not on the local scale, the amount and gross radiative properties of cloud do not depend on space and time variations in the nucleating properties of the atmosphere. If this is so, the cloud problem can, at least in principle, be resolved within the logical framework of the model without further empiricism—the dynamic and thermodynamic processes generate and dissolve the cloud; the condensation and evaporation react grossly on atmospheric temperature and motion. The situation in respect of aerosol is rather different. There are both surface and internal sources of both man-made and natural aerosols. These aerosols are transported by atmospheric motions but there is no first-order feedback between aerosol content and motion—the interaction is slow and through the radiative terms. Transport can be handled by the models—on the small scale this is done in "air pollution models" and on a scale of hundreds of kilometers, R. J. Murgatroyd has recently applied a detailed fore-

casting model (Bushby-Timpson, 10 layer 80 km grid square) to the problem of three-dimensional transport of pollutants. At the time of writing only abstracts of Murgatroyd's work have been seen: the outstanding problem may be the handling, with a tolerable number of layers in the vertical, of the quasi-isentropic nature of the three dimensional motion and the resultant tendency for locally injected pollutant to be confined, in a stable atmosphere, to gently-sloping layers of very limited depth. An approach other than empirical to the specification of a source terms for the natural aerosol (e.g., raising of surface dust by the model-generated wind) is a development for the distant future. Man-made sources must always be empirically specified.

3.4 Future Development of Climatic Models

The brief overview of simple climatic models did not lead us to any very encouraging conclusions. It is not easy to formulate recommendations for further work with them because one cannot have real confidence, even qualitatively, in the results so far announced. On the other hand, there is now general agreement that modeling of the whole surface-atmosphere complex is essential to an understanding of climatic change consequent on any local or global modification of the atmosphere or surface and inactivity will not advance the cause. A possible approach would be use of the comprehensive global weather models of the type developed by Smagorinsky's group in something like a "Monte Carlo technique" mode to accumulate statistics on the climates of atmospheres of differing constitution, but this expensive undertaking is not likely to commend itself to the very few institutions at present able to contemplate it. The expense and elaboration are sufficient to make this a very inefficient approach, so long as the models do not carry cloud cover as a dependent variable.

It would perhaps be useful to continue development of the simple models as "educational toys"—for example: to attempt to improve the treatment of mean meridional circulations in Sellers' model and then examine the effects of imposing different relations between albedo and temperature. This relation is one of the keys to the modeling of long-term climatic change, and it might be that a new approach with more parameterization of dynamical aspects and more detailed treatment of radiation and cloud physics could be explored. The simple models carry parametrization of all processes to the extreme.

4.0 TRANSPORT OF POLLUTANTS IN THE CONTEXT OF LONG-TERM EFFECTS

Short- and medium-range transport of pollutants is treated in the context of urban and regional air pollution models—see for example, the report of a seminar on Urban Air Pollution Models recently organized by NAPCA [24]. Mixing of a long-lived pollutant on a global scale is exemplified in the study of CO_2 by Bolin and Keeling [2]; and work of this kind can be, indeed has been refined, but its impact would be more on general dynamic meteorology than on the study of pollution and its effects. (The pollutant might be used as a tracer of atmospheric motion and as a lead to the parameterization of various scales of motion.) Between the local and the global scales, there is a scale where the transport of pollutants, averaged in the long-term, is of biological interest and could become of political importance. The problem is that of deposition of pollutants and the products of their transformations downwind of major extended sources. Regional or sub-continental pollution models are required for this investigation, and they are not yet developed and tested. The problem is referred to in Section 3 where it is suggested that such a model might be constructed from the more detailed meteorological models now being developed by Weather Services. The most obvious possibility is to include the continuity equation for a pollutant, with appropriate boundary and internal source terms, within a "primitive equation" meteorological model; but difficulties as well as possibilities are obvious and another approach might be more profitable at least initially. The fact that pollution on this scale is an international problem is already recognized: international negotations may at some stage be called for, and it is not too early to try to establish a sound basis for them. "Specification of the internal source term" takes us back, of course, to little known areas of air chemistry so there is no reason to expect easy success. Empirical testing of a model on this scale might well throw some light on the chemical problem. The requirement is to relate the type of fact summarized in Fig. 3—the amount of sulphur deposited in precipitation over the United States—to the natural and artificial sources.

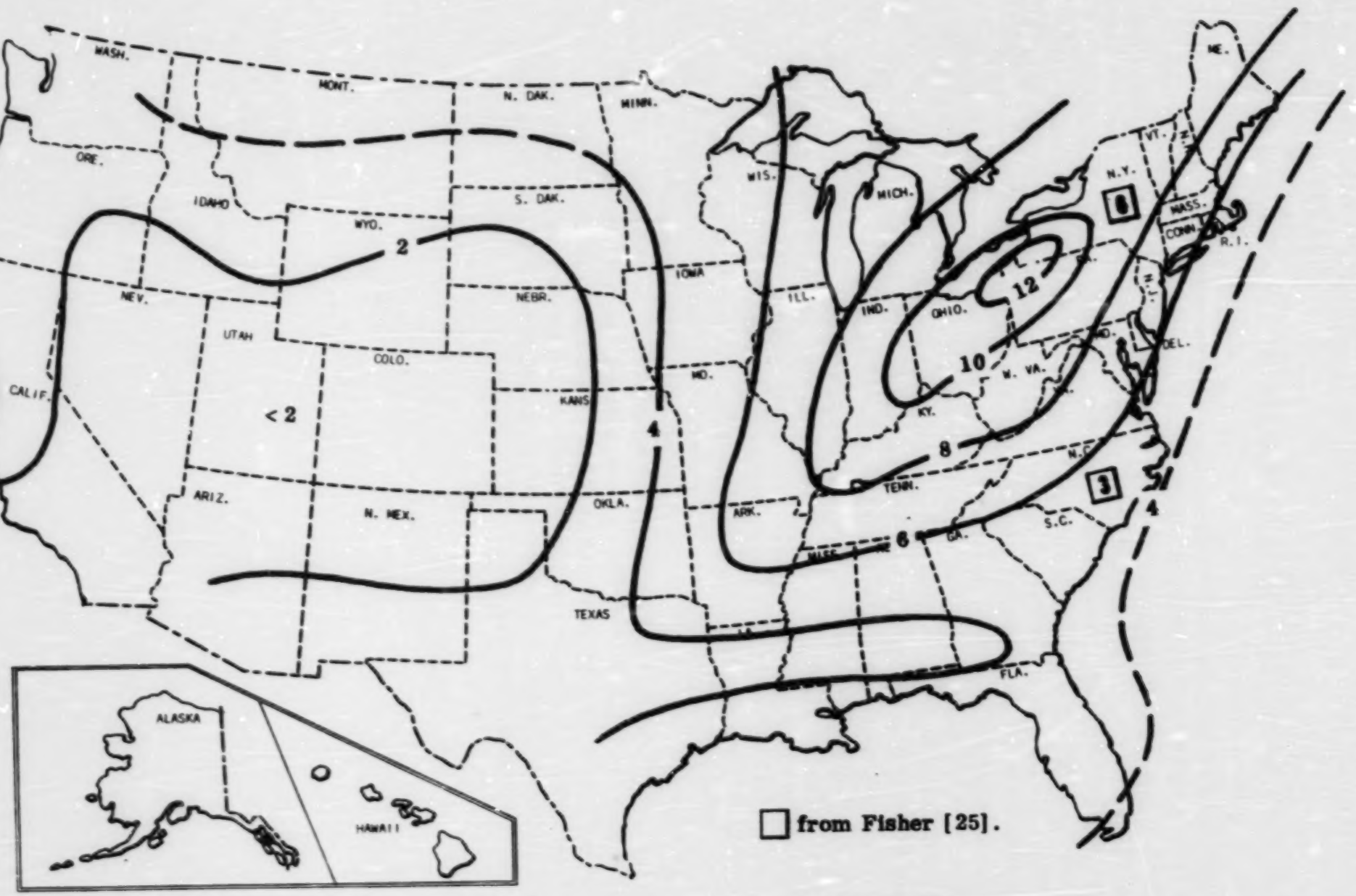


Fig. 3. Sulfur in precipitation over the U.S.

Unit—gm^{-2} yr^{-1}. Isopleths from Ericksson, quoted by Robinson and Robbins.

5.0 POLLUTANTS AND RADIATIVE PROCESSES

The most obvious way in which a pollutant can affect climate in the long term is by interfering with the radiative processes in the atmosphere. We have a fairly sound knowledge of the theory of radiation in the atmosphere and can compute the first stage in the chain of effects which would follow the addition of a pollutant of known optical properties. In this section we examine the relevance of this knowledge to our problem of predicting the long-term effects of pollution. We find major deficiencies in meteorological modeling and in our knowledge of the optical properties of aerosol.

5.1 Radiative Effects of CO_2

CO_2 absorbs solar radiation in the near infra-red in regions where water also absorbs. This is not an important source of atmospheric heating and can be neglected when the effects of change of CO_2 content are considered. Unlike areosol pollution, CO_2 pollution does not change the heat supply immediately available to the planet. There is no reason to expect change of atmospheric CO_2 content to affect the mean temperature of the planet since the outgoing radiation is fully black at some temperature already existing in the system and the total outgoing energy is controlled by the solar input. An increase of CO_2 should not, on this qualitative argument, change the average net terrestrial radiation at either the top of the atmosphere or the earth's surface, and, since an increase of CO_2 could only increase the emissivity of air, a reduced temperature should result high in the atmosphere, compensated by an increased temperature near the earth's surface—the "greenhouse effect."

Quantitative attacks on the problem confirm the qualitative deductions, but over the years there have been varied estimates of the magnitude of the rise of surface temperature which might accompany an increase in CO_2 content. Callendar, before World War II, and Plass in the early 1950's, computed the effect of CO_2 alone, and Plass concluded that doubling the CO_2 content would result in an average surface temperature increase of 3C to 4C. Kaplan argued that the presence of cloud (in a fixed amount equal to the present global average) would reduce this figure by about 60 percent. Kondratiev pointed out the importance of considering the H_2O absorption region. Using a fixed water content and ignoring cloud, he computed surface temperature changes only 15 to 20 percent of those suggested by Plass. All these computations were of the primary reaction of the atmosphere on the supposition that CO_2 content increased and nothing else changed except air temperature. Möller [26] was the first to study a secondary effect influencing both solar and terrestrial radiation. (His paper also contained a succinct summary of earlier work.) He pointed out that increased temperature of the lower atmosphere inevitably implied increased water content. He found, assuming unchanged relative humidity, that this greatly affected the computed temperature change and he quotes a rise in surface temperature of 10C for a doubling of CO_2 content. We are, at this level of complication, concerned with a destabilizing situation—the "greenhouse effect" induces a rise in surface temperature, which evaporates more water, which in turn increases the "greenhouse effect." Möller also, however, pointed out the existence of further secondary effects, particularly those dependent on cloud. His computations, like all previous and subsequent computations, held the amount of cloud constant. Möller

attempted a semi-quantitative estimate of the effects of cloud on the CO_2 problem. He concludes:

> "It is not difficult to infer from these numbers that the variation in the radiation budget from a changed CO_2 concentration can be compensated for completely without any variation in the surface temperature when the cloudiness is increased by—1 percent of its value or—the water vapor content (decreased) by 3 percent of its value. No meteorologist or climatologist would dare to determine the mean cloudiness or the mean water vapor content of the atmosphere with such accuracy, much less can a change of this order in magnitude be proved or its existence be denied."

The studies which culminated in Möller's work proceeded by computation of the downward terrestrial radiation at the earth's surface and related this to the change in surface temperature by consideration of the surface energy budget. Manabe and Wetherald [27] extended the scope of the computations to cover the temperature structure of a whole atmosphere in convective-radiative equilibrium. They considered all radiating gases, with a fixed "climatological" cloud distribution. They specified water content by a given relative humidity, and, as did Möller, included the effect of changed water content on solar radiation. They have, therefore, the same "destabilizing" condition as Möller, but its effects on surface temperature are tempered by convective readjustment and their final result for a 100 percent increase in CO_2 content, with (existing) average cloudiness,is a surface temperature increase of 2.4C. Their computations also showed a considerable temperature decrease in the stratosphere as we would expect from our initial qualitative agrument.

There are some minor aspects of Manabe and Wetherald's paper which might not secure universal acceptance, but it is reasonable to suggest that, with the exception of a rather specialized area mentioned below, their computations are the best that need be made for the effect of a change of CO_2 content on an atmosphere with specified constant cloudiness. Refinement of their treatment of both radiation and convection is possible, but the work would still be open to the (quoted) criticism which Möller made of his own work. It seems reasonable to argue that an increase in atmospheric CO_2 content would result in an initial tendency for warming at the surface and cooling in the high stratosphere but there will be no justification for a forecast of the final equilibrium temperatures until we have made an order-of-magnitude advance in the complexity of atmospheric models, to include the distribution (and ideally also the albedo) of cloud as a variable.

The aspect in which further computation might at present be rewarding concerns the effect of CO_2 concentration change in the mesophere. At the upper limit of Manabe and Wetherald's computation (around 40 km or 2.5 mb) doubling the CO_2 content from its current value leads to a temperature decrease of about 10C. At about this height computations begin to be complicated by the fact that CO_2 is not in local thermodynamic equilibrium. It is, however, much the most effective radiator in the atmosphere at these heights and temperatures. The temperature and composition of the air in these regions are linked through primary photochemical processes and secondary reactions, some of which are sensitively temperature-dependent. The region of concern spreads

through the temperature maximum and into the lower ionosphere where properties important for radio propagation might be changed. An investigation of the consequences of increased CO_2 content in this region of the atmosphere would probably be rewarding. To the first order, it would be acceptable to hold tropospheric conditions constant.

5.2 Radiative Effects of Atmospheric Aerosol

Addition of a purely scattering material to the earth's atmosphere necessarily increases the earth's albedo. The amount of solar radiation absorbed is reduced, the re-radiated energy is reduced and the effective emissivity is not changed so the effective radiative temperature must decrease. The detailed effect on surface temperature is not of course obvious, but it is reasonable to expect a cooling on balance. If, however, the added material absorbs as well as scatters solar radiation, it may not increase the planetary albedo; if it absorbs terrestrial radiation it may tend to increase surface temperatures by the "greenhouse effect," even though the overall mean planetary temperature is reduced. M. Atwater has made some computations indicating the magnitude of these effects which are summarized here: details are given in Appendix 3.

It seemed useful to investigate an extreme but not inconceivable degree of aerosol pollution, and because of availability of programs a near-surface rather than a stratospheric aerosol layer was considered: a layer 300 m thick containing 10^6 particles cm^{-3} distributed as—

$$n(r) = 5.635 + 10^{18}\, r^3 \exp(-45.1272 r^{0.25}) \quad (r \text{ in } \mu m)$$

(which has a maximum concentration at radius 0.005 μm) is consistent with the distributions investigated by Petersen, Paulus and Foley [28]. (It reproduces closely the distribution shown in Fig. 6 of their paper.) This mode radius is, of course, very small for heavy near-surface pollution, but the properties of the layer as a whole are not unrealistic and since the accent is on long-term effects, it is logical to postulate a persistent aerosol. For a refractive index of $\nu = 1.1 - 0.1i$ for solar radiation and $\nu = 1.1 - 0.25i$ for infra-red radiation*, we find for solar radiation—

Absorption coefficient	σ abs $\sim 1.0\ km^{-1}$
Scattering coefficient	σ scat $\sim 0.5\ km^{-1}$

and for terrestrial radiation—

$$\sigma \text{ abs} \sim 0.1\ km^{-1}$$
$$\sigma \text{ scat} \sim 0.002\ km^{-1}$$

These properties correspond to a horizontal visibility of 2.5 km and an Ångström turbidity coefficient for the layer of 0.2. The absorption coefficient is considerably larger (about × 5) than that observed in an extensive haze layer about 600 m thick over southern England (Robinson [29]) but less than some measured by Waldram [30] in "heavy industrial haze." On a cloudless summer day in mid-latitudes the

*These refractive indexes were chosen to produce appropriate bulk optical properties of the aerosol layer—see M. Atwater, Ph.D. Thesis, New York University.

added absorption of solar radiation by this layer corresponds to a heating-rate for a few hours around noon of about 50K per day. The heat concerned is, of course, lost to the surface: the total effect of the aerosol on the lower atmosphere is to decrease the availability of solar radiation and over a strongly absorbing surface to decrease the actual heating of the atmosphere. The computation of extra cooling by thermal radiation depends greatly on the air and surface temperatures assumed, but a reasonable figure is 2 to 3 K per day over the layer as a whole, with warming near the surface. With this particular aerosol over an absorbing surface, climatic effects depending on long-wave radiation are considerably less than those depending on solar radiation in most summer latitudes, but the long-wave radiation effects dominate in winter.

It is obvious that if we are concerned with an aerosol which absorbs but does not scatter, or with an absorbing and scattering aerosol over a high albedo surface, the result will be a decreased global albedo. Atwater's computations (Appendix 3) investigate this effect for a layer 275 m thick with various scattering and absorption coefficients and various surface albedos, with some indication of the effect of zenith angle and the ratio of forward to backward scatter. For example, the aerosol layer considered above increases global albedo when the surface albedo is less than about 0.2, but decreases global albedo for larger surface albedos. This is a case of extremely high aerosol absorption but the computations show, for example that most industrial aerosols would reduce global albedo over a snow or ice surface with the low solar elevations of sub-polar localities.

Measurement of the radiative effects of "pollutant" aerosol must take place against the background of the "natural" aerosol which we have seen (Section 2.2 and Appendix 1) to be more prevalent than "pollutant" aerosol over a large part of the world. Lettau and Lettau [31], for example, have shown that attenuation (though not absorption) of direct solar radiation by aerosol at a desert location in Peru is comparable with that measured in the suburbs of London, England, before operation there of the Clean Air Act; and during the recent "Bomex" observations in the western tropical Atlantic layers of dust, presumably of Saharan origin, were observed to absorb several percent of the incident solar radiation (A. J. Drummond, personal communication).

Comparison of observed sky radiation in relatively unpolluted localities with that computed for molecular scattering suggests that aerosol commonly scatters downward 5 to 15 percent of the incident solar radiation. In apparently unpolluted air over the English Channel, both Roach [32] and Robinson [29] observed an absorption of solar radiation between 10,000 and 20,000 ft about 3 percent in excess of that expected from gaseous constitutents, with an excess upward scatter of about 1 percent. More data of this type can be expected from the "Bomex" experiment. On the other hand, some records of diffuse sky radiation taken on the Antarctic continent can be explained in terms of molecular scattering only.

Light scattering observations—searchlight, laser and twilight—suggest that the stratospheric particulates in the Junge layer scatter about the same amount of visible radiation as does the clear air at the same height. Stratospheric scattering of the total solar radiation appears to be significantly in excess of the Rayleigh value at all

heights. Stratospheric scattering is too small an effect in absolute terms to be deduced with confidence from the surface measurements of solar radiation normally made by meteorological services, but it certainly appears that some Antarctic air masses have a lower tropospheric concentration of aerosol than most stratospheric air.

Observations of a secular increase in atmospheric turbidity (a measure of extinction of solar radiation in excess of that to be expected from a clean atmosphere) at Washington, D.C. and Davos, Switzerland led McCormick and Ludwig [33] to raise the whole question of long-term effects of pollutant aerosol. (Washington and Davos are probably the only localities with really reliable records over a 50-year span: Davos is an alpine station far removed from major pollution sources). Flowers, McCormick and Kurfis [34] have examined the results of a network of stations measuring turbidity and have established the pattern shown in Fig. 4 for non-urban stations within the U.S. They appear to have established a simple and reliable indicator of total aerosol content—natural and man-made—valuable as a long-term monitor.

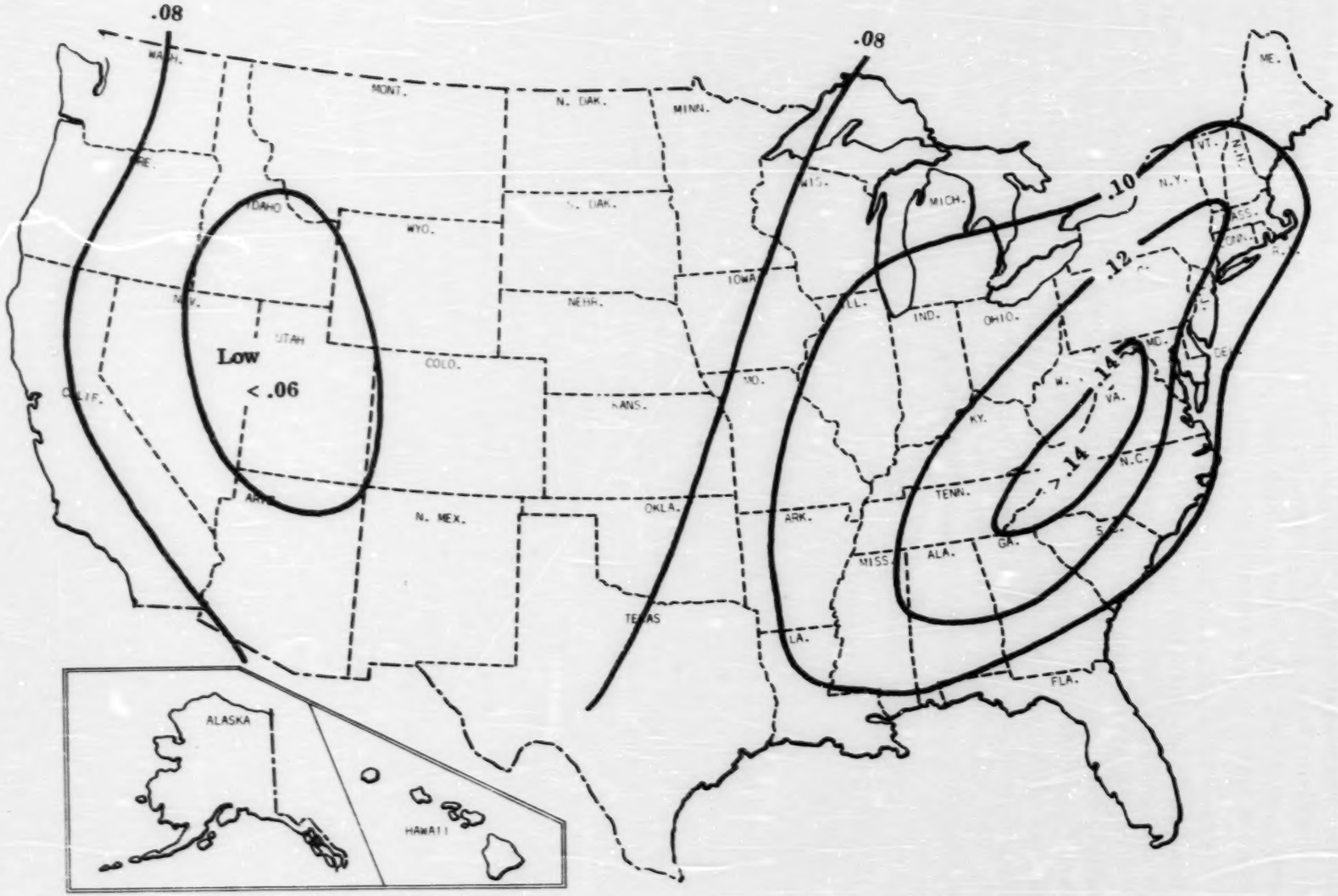


Fig. 4. Annual mean of turbidity over the United States excluding urban stations 1961—1966 (Flowers, McCormick and Kurfis).

6.0 POLLUTANTS AND CLOUD CONDENSATION PROCESSES

An understanding of the physical processes involved in nucleation and an appreciation of the properties of natural nucleus populations is essential to a judgment of the likelihood of modification of precipitation processes by pollutant aerosol. On the other hand, there are broad thermodynamic arguments which suggest that conceivable levels of pollution are not likely to affect climate on a global scale by way of nucleation effects alone. To avoid an unbalanced accumulation of detail, only a brief overall description of the nucleation process is included in the body of this report: the necessarily much fuller treatment by Dr. T. B. Smith and his colleagues is in Appendix 1.

There is much more likelihood of detectable, even serious, modification of precipitation processes on a local and perhaps on a regional scale, and some of Dr. Smith's conclusions on these matters are mentioned here and repeated in Appendix 1.

6.1 General Nature of the Nucleation Process

Certain atmospheric particulates act as cloud condensation nuclei (CCN) or ice nuclei (IN). CCN are those particles in the atmosphere on which water vapor condenses to form cloud droplets. IN are those particles which have the special property of nucleating the ice phase in clouds either by nucleating supercooled droplets or by serving as centers upon which ice is deposited directly from the vapor phase.

The concentration in the air of those CCN which are active at the maximum supersaturation existing in a cloud determines the concentration of cloud droplets. This is one factor controlling the efficiency with which cloud droplets can grow by coalescence to form raindrops in a warm cloud. Thus, if the concentration of effective CCN (and therefore cloud droplets) is high the average size of the droplets will be small and their growth to raindrop size will be difficult. This is thought to be the situation for clouds forming in continental interiors. On the other hand, in maritime air masses the concentration of CCN is quite small, the average size of the cloud droplets is therefore larger than in continental clouds, and raindrops are produced more readily. The addition of CCN to a cloud mass therefore increases the time required for the development of precipitation. On the other hand, if relatively small numbers of highly efficient CCN (so called giant nuclei) are introduced into a cloud they may serve as preferential centers for condensation and these droplets may increase rapidly in size to form raindrops. (This, of course, is the principle behind the seeding of warm clouds with giant salt particles in order to enhance precipitation.)

In the case of clouds which extend above the 0°C level, the growth of ice particles by the Bergeron-Findeisen process (distillation from supercooled droplets to ice particles) provides another mechanism by which precipitation particles may be formed. The concentration of ice particles is related to the concentration of active ice nuclei in the cloud. Below a certain critical concentration of ice nuclei, the ice particles can grow to precipitation size fairly readily. However, if the ice nuclei exceed this critical concentration the formation of precipitation may be hindered. The nucleation of the ice phase in clouds has an additional and important effect, namely, that it releases a significant quantity of latent heat which increases the buoyancy of the cloud. Under certain environmental conditions, this can result in the

"explosive" growth of the cloud. These ideas have received general confirmation in experiments in which supercooled clouds are seeded with artificial ice nuclei.

6.2 Nature and Origins of Cloud Nuclei

Since the maximum supersaturations which exist in clouds rarely exceed 1 percent, only the larger (say > 0.05 μm) and generally hygroscopic particles in the air act as CCN. Typical concentrations of CCN are 10^2 cm^{-3}, whereas the total concentration of particles may be of the order 10^5 cm^{-3}. The surface of the earth and the ocean are thought to be sources of CCN. Recently, certain industries (e.g., paper mills) and artificial and natural fires have been identified as sources of very effective CCN.

Natural ice nuclei are very rare. Typically, only 1 particle in 10^{11} in the atmosphere is effective as an ice nucleus at a temperature of -10°C. The exact mode of action of an ice nucleus is still a matter of dispute. The relatively few studies which have been made of the composition of natural ice nuclei indicate that silicate minerals from the earth's surface are dominant. Certain industries (e.g., steel mills) emit large quantities of ice nuclei into the atmosphere. Measurements made at three widely separated sites (Hawaii, Alaska, Washington) indicate that under certain conditions ice nuclei may be advected over distances of thousands of miles.

6.3 Pollution and Nucleation—Condensation Nuclei

It is estimated that globally only a few percent of the CCN are man-made, but in localized urban areas the number of artificial nuclei may exceed the natural population. Pollution aerosol may also play a part in the activation or de-activation of nuclei by coagulation with natural nuclei. Pollutants may produce nuclei by secondary reactions in the atmosphere.

The effectiveness of additional CCN in modifying precipitation processes depends in practice more on their size range and that of the natural population to which they are added than on overall numbers. They change the rate at which the coagulation process becomes effective in producing rain, not the nature of the process. Some computations in realistic cases are set out in Appendix 1 where it is concluded that, "the results can be viewed from the perspective of the overall mechanism involved in the modification of the coalescence process. It is generally assumed, to a first approximation, that the dynamics and lifetime of the warm cloud are not affected by changes in the coalescence growth rate. This means that precipitation changes can result only from changes in the storage of cloud water, either increased or decreased. Since the water supply of most of the large clouds exceeds the cloud storage capacity under natural conditions, it is only the smaller and marginal clouds which are likely to be affected by the changes in particulate concentrations."

6.4 Pollution and Nucleation—Ice Nuclei

We have seen that natural ice nuclei are not abundant; it also appears that very few pollutant particulates possess the ice-nucleating property. Details of

some measurements are given in Appendix 1 which suggest that urban (specifically Los Angeles) air may sometimes contain fewer ice nuclei than the surrounding rural air—presumably because of deactivation by coagulation. On the other hand, large numbers of IN have been detected in the effluent from steel mills, and some urban atmospheres (specifically Seattle) have been found to contain an excess of IN. Two further possible sources of pollutant IN are examined in Appendix 1, where it is suggested that they are probably not significant. The first, suggested by Schaeffer, results from reactions between lead compounds emitted in automobile exhaust gases and iodine vapor, to produce PbI, which is a highly efficient IN. The second is the exhaust particulates of jet aircraft.

Direct measurement within the exhaust of a jet engine detected changes in ice nucleus content which were not large enough to be considered significant, and the importance of Schaeffer's mechanism has not been confirmed by independent observation in the atmosphere, perhaps because iodine vapor, even in the minimal concentrations which would be significant, exists only as a rare local pollutant. To quote from Appendix 1, "The foregoing studies indicate that the effect of air pollution on ice nuclei concentrations may be quite variable, depending on the type of pollution, concentrations, etc. Values may range from the 1000/liter found in the French industrial regions to changes of less than an order of magnitude to, finally, the Los Angeles area where there is a tendency for lowest ice nuclei values to be associated with heavy pollution. Schaefer's comments on iodine and lead reactions in automobile exhausts can be viewed in the perspective of the measurements made in the two metropolitan areas of Seattle and Los Angeles. In the one case, area concentrations may have been increased by a factor of six as a result of automobile and/or industrial sources. In the other case, with industrial sources more restricted than in most areas, the ice nuclei counts in pollution tend to be lower than in unpolluted regions. These comments support the general conclusion that local effects of the iodine and lead reaction may occur but there appears to be no substantial evidence of a widespread contribution from this source.

Long-term records of ice nuclei variations essentially do not exist. Partly this results from the modifications and improvements in ice nuclei measurements which have occurred in the past 15 years. Grant at Colorado State University, however has measured ice nuclei routinely at Climax, Colorado, for the past 10 years. Fortunately, these measurements have been consistent in terms of observational technique. Grant (private communication) reports no definable change in ice nuclei counts over the 10-year period which might be considered as a background trend. It remains possible, however, that areas such as the eastern sections of the United States or portions of industrial Europe might be experiencing gradual increases in ice unclei content but that an efficient natural removal process might serve to make the trend indiscernible at Climax. The Climax data also fail to show any pronounced influence from polluted areas to the west such as Los Angeles. This is in keeping with measurements made in the Los Angeles area itself which fail to show widespread ice nuclei effects, even in the source region itself."

6.5 Observational Evidence of Precipitation Changes

6.5.1 Local Effects

Substantial evidence of precipitation modification is meager but significant. The two outstanding and oft-quoted examples are an increase in reported rainfall at La Porte, Indiana (Changnon [35]), and local decreases in Australia (Warner [36]). Changes in precipitation of 25 to 30 percent have been found in these areas. Hobbs, Radke, and Shumway [37] provide suggestive data relating precipitation increases in Washington to the location of major industrial complexes. Increases of over 30 percent were found for the period 1947—1966 compared to 1929—1946. Miller [38] found apparent increases in precipitation of the order of 15 percent over Long Island and downwind of New York City. The remaining examples of precipitation effects have been summarized by Changnon [39] and Peterson [40] and generally show changes in precipitation of the order of 10 percent or less. It is useful to examine these cases in the perspective of the preceding sections.

Changnon [39] has found that much of the increase in reported precipitation at La Porte occurs during the warm season and that the number of thunderstorm days is increased significantly and concludes that midwest urban areas produce significant increases in convective activity. Principal precipitation effects come from an increase in the number of days with 0.25 inch of rain or more. The conclusion was reached that thermal and frictional effects were primarily responsible for the urban effects in the midwest and that La Porte represented a unique situation due to an unusual combination of urban, industrial, and lake contributions.

These conclusions are in agreement with the available results from advertent modification experiments. Increases in precipitation of as much as 30 percent in the annual rainfall in an area such as La Porte seems to require thermodynamic seeding effects, i.e., through the release of additional convective activity. Microphysical effects such as have been observed at Climax should appear primarily in terms of increased number of light precipitation days and should, as well, be more related to stratiform cloud types. Ice nuclei, according to Langer [41] were measured in concentrations of about 30/liter at -20°C. Results of seeding programs at Flagstaff and elsewhere suggest that this concentration may not be sufficent to produce the dynamic seeding effects found in the advertent cumulus seeding programs. Condensation nuclei effects, although not discussed at length in the La Porte example, have not been shown to result in such large precipitation increases. It is concluded that the thermal and frictional effects, resulting in frequent updraft regions in a localized area and the stimulation of convective motions in this area, are the most likely causes of the pronounced increases indicated by the La Porte observations. A corollary of this conclusion is that it should be possible to identify those days on which convective activity is so poised that additional stimulation is sufficient to release the latent convective instability in the preferred area. This identification problem has been considered in the case of cumulus seeding with considerable success.

The Australian study by Warner has shown apparent decreases in precipitation of the order of 25 percent. These effects have been attributed to the production of large numbers of small cloud droplets and a consequent decrease in coalescence

growth rate. Cloud droplet concentrations averaged about 900 per cm^3 in the inland areas where cane smoke might have affected the cloud microstructure. Peak droplet concentrations were over 2500 per cm^3.

It has been indicated earlier that the principal effect of a slowdown in the coalescence growth rate would be to prevent precipitation from developing in the smaller clouds whose lifetimes are not particularly long. It has been estimated that a decrease of 25 percent in total precipitation would result if precipitation were eliminated from clouds with diameters less than 3 to 4 km. Larger clouds (with longer lifetimes) might produce rainfall in much the same manner, regardless of cloud droplet concentration. There is evidence in Warner's data that the number of showery days has been decreased substantially but that the amount of rain per shower day has not changed in a similar manner.

The concentration of cloud droplets reported by Warner is not extreme in comparison to numbers found in other areas. In Flagstaff, concentrations of 700—1000/cm^3 are frequent and occasional values of 2000/cm^3 are observed. In Flagstaff, however, substantial coalescence rain occurs in clouds of 3 to 4 km diameter in spite of the large droplet concentrations. Number of droplets/cm^3 should not be the ultimate predictor of the occurrence of coalescence precipitation, and it is entirely possible that a sufficient number of large particles may have been present at Flagstaff to initiate the precipitation. It is concluded, therefore, that the Australian data, if statistically sound, may show the result of added condensation nuclei but that a droplet distribution devoid of large particles would be required to produce such a marked change in precipitation as has been reported.

Hobbs, Radke, and Shumway [37] indicate that centers of increased precipitation appear downwind of local industrial sources such as smelters or paper mills. Precipitation increases are quoted at 30 percent for several areas when comparing the industrially active, recent years (1947—1966) with the earlier years of 1929—1949. The authors point out, however, that the entire northwest apparently experienced heavier rain in the 1947—1966 period than in the earlier era. The value of 30 percent includes both the effects of the wetter areal trend and the possible effects of industrial sources. No attempt has been made to separate these two effects and a cursory glance at the data suggests that no more than a 15 to 20 percent increase should be attributed to the industrial effects. The authors attribute the apparent increase to the effects of condensation nuclei released from the industrial plants.

Observations are described in the article by Hobbs et al, which indicate numerous occasions of cumulus cloud development downwind of the particular plants in question. On one occasion, a cloud street of 30 km in length was reported downwind of a paper mill in a manner which can hardly be attributed to condensation nuclei. Evidence is also given to indicate that the precipitation in the 1947—1966 period was more convectively generated than in the earlier period. The size of the apparent increase (second in magnitude only to that reported from La Porte), the cloud observations, and the convective nature of the precipitation all suggests that dynamic effects on the precipitation mechanisms should not be ruled out of consideration. The similarities to the La Porte situation in terms of cloud developments and precipitation increases are striking. In addition, calculations such as have been possible to date do not support

such large precipitation increases as 15 to 20 percent (in annual rainfall) as a result of stimulation of the condensation-coalescence process. It is concluded, therefore, that the Washington data may afford another example of a thermodynamic or frictional effect on precipitation in an environment somewhat different from that existing at La Porte.

Data on Long Island (Miller [38]) suggest precipitation increases of around 15 percent downwind of New York City compared to surrounding areas. No detailed examination of the concurrent environment conditions has been given. The pattern of the increase, with isohyets symmetrical with respect to the island's longest axis, are suggestive of dynamic effects rather than microphysical causes. It could be hypothesized that convection over the island with reference to the cooler, surrounding water surfaces might contribute to such an isohyetal pattern. As in the La Porte case, the effect of New York City could be to initiate the convection in a consistent location.

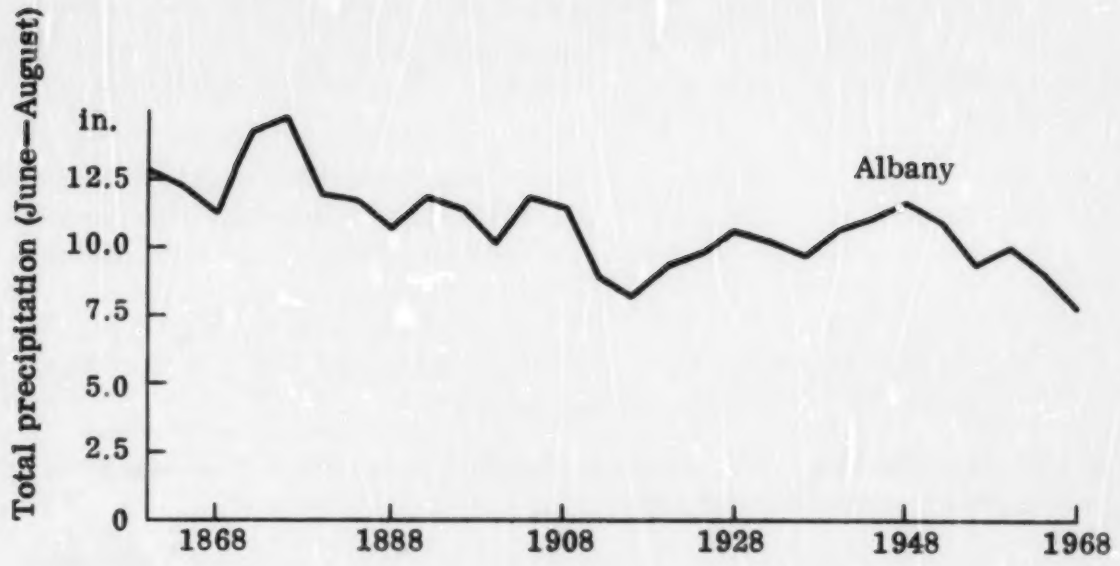
The remaining examples of precipitation changes are of the order of 10 percent or less. Little physical documentation of the environment conditions is usually given and the increases themselves are small enough to be more subject to doubt than the preceding cases. Under these conditions, it is possible to comment on plausible reasons for the precipitation effects if, in fact, they can be shown to be significant.

6.5.2 Regional Effects

In view of the possible influence of nuclei on warm cloud precipitation, as shown by the Australian data, it was decided that long-term trends in summer rainfall in several areas of the United States should be examined. In the light of probable increases in pollution levels over a period of years in the eastern sections of the country, widespread effects on precipitation might be expected to appear first in summer precipitation amounts in these areas. June, July, August rainfall was plotted for a period of years for several stations in the eastern part of the United States. Data for Albany and St. Louis are shown in Fig. 5. Data for Nashville, Cincinnati, and Philadelphia were also examined but show no particular trends over the past 100 years or so.

The most pronounced summer rainfall trend found at any of the five stations is shown in the curve for Albany in Fig. 5. Summer precipitation in this area appears to have declined 20 to 25 percent over the period of the last 100 years. It can be argued that this trend was particularly pronounced during the past 20 years, but such short-term trends may occur as a result of a number of causes. At St. Louis, the general trend was downward until the mid-1930's, after which evidence of rising summer precipitation amounts is apparent.

Long-term trends in areal precipitation amounts could result from a variety of causes. Along with the possibilities of nuclei and moisture effects, it is quite probable that true climatic trends, identified with long-term circulation changes, may also play a role in determining precipitation trends. As we have seen such long-term changes may be associated with environmental changes, or may be a result of "almost-intransitivity." It is important, therefore, that identification of such areas as Albany be followed by more detailed studies to identify possible causes.



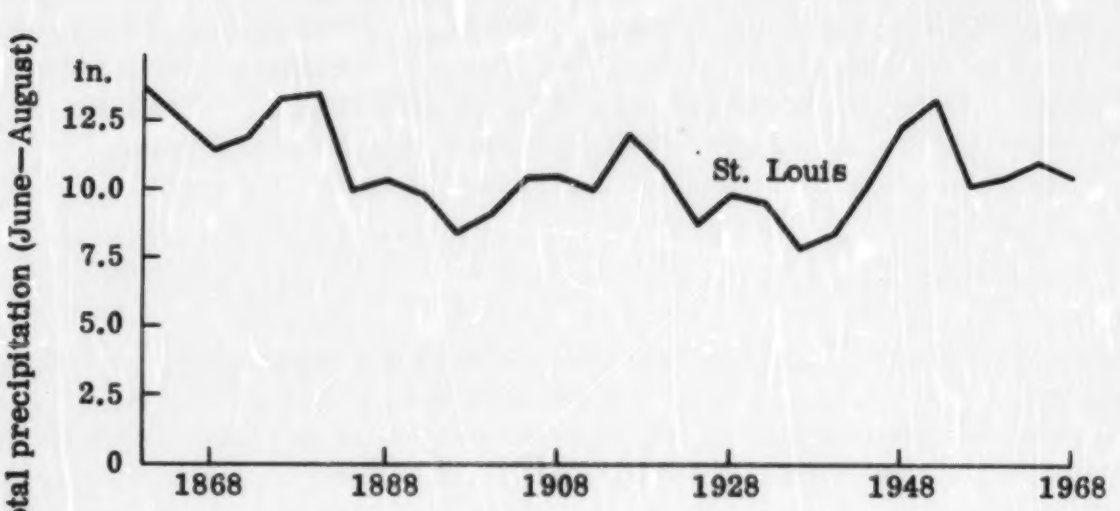


Fig. 5. June—August precipitation trends.

7.0 THE BIOSPHERE AND LONG-TERM EFFECTS OF AIR POLLUTION

The long-term effects of air pollution on the biota of the earth are subtle, very poorly understood, and presently receiving only very limited attention. These conclusions have been reached after a review of recent literature and research proposals and personal contact with many of the leading investigators in the fields of air pollution investigation.

In general, we need more meteorological and chemical information regarding the fate of air contaminants. As suggested in Table I of Appendix 2 the terminal nature of numerous gaseous pollutants is a chemical different from the source nature of the pollutant. What are the terminal forms of oxides of sulfur, carbon and nitrogen? What are the terminal forms of organic primary and secondary (photochemical) pollutants? Where are they deposited (over land, oceans)? How are they deposited (rain, dust)? In what quantities are they deposited?

Awareness of the fate (compositional, spatial, temporal) of air contaminants will suggest the nature of potential problems.

a. Input of significant amounts of nitrates and nitrites (conversion products of nitrogen oxides) into certain ecosystems might result in or contribute to eutrophication of certain water resources.

b. Input of sulfates (conversion products of sulfur oxides), if large enough (specific levels unknown and would vary depending on soil characteristics), could influence soil pH and stimulate the microbial formation of sulfides. Significant alterations in pH would profoundly effect nutrient availability to plants and plant disease caused by certain soil pathogens. Sulfide increases would accelerate corrosion potential of the soil.

c. Fate of oxides of carbon is extremely unclear. Many questions exist regarding the global balances of CO_2. Are plants growing better in response to increased availability of CO_2? Are temperatures being altered? In the case of CO, the nature of the subtraction process remains unknown. It may be microbial or photochemical. If microbial, what microbes are important? In what ecosystem do they operate? Are they under stress from any other environmental alterations?

Knowledge of the potential problems will permit the design of experiments, employing plants and animals, capable of assessing the impact and monitoring the influence.

More meteorological information is required with respect to the influence of air pollutants on climatic and weather parameters.

a. What alterations are manifest in patterns of solar radiation? Because of wavelength specificity, the photosynthetic capacity of plants may not be significantly reduced. The germicidal effect of light, however, may be altered as meaningful subtractions may occur in the UV wavelengths. What is the significance of this subtraction on the ecology of airborne spores and other inocula of animal and plant disease agents?

b. What is the significance of particulate pollution in regard to rain and fog patterns? Changes in frequency and intensity of precipitation patterns can have very significant effects on physiological and pathological phenomena of plants growing in urban environments.

Plants currently being used to assess the presence of air pollutants are primarily employed to monitor for relatively large amounts of air contaminants (source; nature; form) and are evaluated by observing acute effects. Plants presently employed in these programs are not used to measure chronic or long-term influences. Vegetative monitoring of long-term influences may require (if it is at all possible) different plants which will react to the pollutant in its terminal form and which can respond to relatively low levels. There is at least one very significant exception to the above generalizations. In the case of accumulatory materials such as lead, fluroide and chloride, plants may provide very convenient and accurate monitoring systems. They do not appear to be used currently in this capacity.

In summary, the biological significance of the long-term influence of air pollutants cannot be evaluated without greater information with regard to the physical and chemical characteristics of the environment and environmental contaminants as outlined above.

8.0 MONITORING

8.1 Rationale of Monitoring Long-term and Large-scale Phenomena

Monitoring is described in Section 1.3 as the "purposeful, controlled, continuing observation on a global or local scale of a pollutant or of an established or suspected effect of a pollutant." Recent experience has suggested that everyone has his own conception of monitoring and that some expansion of the idea of monitoring which is adopted in this report is necessary. Here, monitoring implies selection as well as purpose. We do not "monitor" a system by continuous watch on all its detail, but by choosing some aspect as an indicator of the state of the system and observing this aspect with the precision, frequency and geographical coverage which is necessary—and just as important with no more effort or coverage than is necessary. The speculative collection of data or the collection of data for occasional use—for example, for the preparation of tomorrow's weather forecast—or for a limited research project is not monitoring. For example (and it is not put forward as a sensible example), the chemical determination of cyanide in river water, either because it might prove interesting or specifically for the purpose of correlation with the respiration rate of fish would not be considered "monitoring." On the other hand, systematic observations of the respiration rate of fish might be described as monitoring cyanide if the correlation were specific and the procedure more sensitive, accurate, universal and/or more economical than chemical methods. If, however, cyanide in the water had no observable effect, or no known potential effect, other than that on the respiration rate of fish, it would be an unnecessary complication to describe measuring the respiration rate of fish as monitoring cyanide in the water. The essence of monitoring is selection—selection of critical phenomena to be watched, selection of observables critical to one or more of these phenomena, and selection and standardization of methods of observation.

Our present task is to watch for long-term changes in climate induced by air pollution and the accompanying changes in biota. Maintenance of a climatic record is the task of weather services: a continually more detailed record is kept, but if it should show changes we have seen that these cannot necessarily be attributed to air pollution. We must, therefore, identify pollutants which could conceivably influence climate in the long-term, study their concentration and distribution and study the problem of monitoring them. We have identified CO_2 and aerosol. CO_2 has so long a residence time that it is almost uniformly mixed. We can handle the minor variations by choice of site and monitor by direct measurement in a few localities. We perhaps do not have sufficient information on the minor diurnal and annual variations at chosen sites to monitor on any but a quasi-continuous basis at present, but we can envisage the situation when a limited number of determinations each year at each site will be sufficient to monitor an established temporal trend. The need then will be for an exactly reproducible, preferably absolute, method, so that if we can establish such a method now we should do so, even if it is more elaborate than less readily standardized methods.

The problems with aerosol are very different. The particulate material is highly inhomogeneous; it (or most of it) has a short life in the atmosphere and in any one locality its incidence is as variable as the weather. Comprehensive global

observation of the nature of atmospheric aerosol would be an enormous task. To reduce the task, we inquire how aerosol reacts on climate, and find it does so through modification of condensation processes and modification of the radiation field. We find that two specialized types of particle are involved in condensation processes, and, perhaps because of the short lifetime, we can identify a fairly constant "natural" background. Study of long-term global effects requires the monitoring of this background, a problem similar to the monitoring of CO_2. Study of long-term localized climatic effects requires the monitoring, on a local basis, of the contribution of local sources.

The radiative effects of aerosol can be studied by the bulk effect of the total atmospheric content on solar and terrestrial radiation. Radiation measurements might be considered a task of weather services, but are at present only partially accepted as such. All types of particles are now involved and no "natural background" fairly constant in time and space, has yet been established. The problem could, therefore, be approached by a broad quantitative survey of one particular radiative effect, which need not in itself be more than an indicator of radiative interference by aerosol, simultaneously with the start of a program involving a limited number of more comprehensive radiation measurements of higher precision selected with reference to our current limited knowledge of the effects and distribution of aerosol. We would be fortunate if the first attempt produced the optimum pattern of measurement for a long-term monitoring system, and it is, therefore, essential that it should be so well documented that its results could be accurately compared in some way with those of future systems.

In the area of biological effects, we are in an even more rudimentary state of knowledge—"The long-term effects of air pollution on the biota of the earth are subtle, very poorly understood, and presently receiving only very limited attention" (Section 7). Research is required rather than monitoring. But this research almost certainly involves monitoring of concentration and rates of deposition of certain pollutants in and on chosen ecosystems, just as research on the economic and medical effects of pollution in cities calls for the monitoring of concentration and deposition rates in which NAPCA is now engaged. As knowledge of long-term effects on biota is increased, it may be possible to limit in quantity, and necessary to improve in quality, the measurements involved. Simultaneously, the possibility will develop of using one or more biological effects as an indicator or a warning of the likelihood of others.

8.2 Artificial Earth Satellites and the Monitoring of Long-term Trends

Artificial earth satellites offer the obvious advantage of global coverage of observations, and the existence of operational meteorological satellites and of experimental earth resources satellites makes it appropriate to consider their possibilities for monitoring pollutants and their long-term effects. A comprehensive study of the possibilities of detection of pollutants from satellite platforms has recently been made (C. B. Ludwig, R. Bartle and M. Griggs [42]). This study concerned itself in the main with currently available techniques and reading it leaves the impression that detection of most pollutants is just feasible but that quantitative assessment with any useful precision is a little beyond current techniques. Further development of on-board techniques is clearly possible, and a more imaginative and

purposeful deployment of satellite vehicles might ease the problems. However, once one gets away from the concept of a space-sharing general-user satellite to consider a system specifically designed for pollution detection, the cost becomes very high indeed and cost-benefit becomes essential.

In the context of the study of long-term effects of pollution, we are concerned with changes in concentration of fractions of one percent per year, and there is no obvious immediate appeal in satellite methods of detection of pollutants, even allowing for the global coverage. Identifying the two pollutants of major interest, CO_2 and aerosol, it does not at present seem that satellite techniques will in the next few years offer any advantage over surface monitoring for CO_2, but that they may make a valuable contribution—indeed may prove essential—to the study of the radiative effects of aerosol. The albedo of the planet earth and its secular changes can be measured most conveniently and directly, if not exclusively, by satellite techniques. Current techniques are not sufficiently precise to allow long-term monitoring. They have been developed in the context of operational meteorological satellites and make a local measurement of albedo; a quantity varying rapidly in space and time, from which the long-term global mean values we require must be deduced. We cannot really assess the value to our problem of current work until we have some feel for the year-to-year statistical variations of global albedo as deduced from these measurements. No space experiment or projected experiment specifically designed to measure the planetary albedo with high (fractions of 1 percent) precision has been publicised. It would call for very high orbits, precision radiometry, and much patience; and it would be very expensive.

The contribution of aerosol to the outgoing terrestrial radiation is required for a full understanding of its long-term effects. It might be most readily detected in the 10—12 μm window, and study of existing IRIS interferograms might give some feeling for possibilities. As has been suggested elsewhere, the requirements of long-term monitoring of particulates and CO_2 should be kept in mind when operational equipment for remote temperature sensing is being designed.

9.0 CONCLUSIONS—AREAS OF UNCERTAINTY

9.1 The Major Pollutants

9.1.1 Carbon Dioxide—Concentration and Life Cycle

The evidence of a global increase of CO_2 concentration appears conclusive, but the rate of increase is still in doubt. The atmosphere-ocean-biosphere partition of CO_2 is not fully understood (see Section 2.3).

Global monitoring is required, hopefully in an international program: A few stations in carefully chosen situations should suffice. Combustion and vegetative growth must be considered in choosing the stations—if their effects cannot be eliminated they must be understood. Available methods of monitoring are sound but not ideal (see Section 8).

Theoretical and observational research on the partition of CO_2 should be encouraged. Isotope (C_{14}) methods are indicated (see Sections 2.3, 8).

9.1.2 Aerosol—Concentration, Nature and Life Cycle

There is evidence of local and regional increases in gross aerosol content of the atmosphere but not yet sound evidence of a global increase. To understand the radiative effects of this aerosol the number concentration, size distribution, composition and refractive indexes of the particles must be known (see Sections 2.2, 5.2). More observations of these details are required—these need not take the form of a regular monitoring program (see below) but extensive tropospheric and stratospheric sampling should continue (see Section 8).

Residence time and removal processes are important areas of research. Quantitative studies of large-scale removal—air mass cleansing processes—are lacking (see Section 2.2).

9.1.3 Sulphur Dioxide—Transport and Life Cycle

The extent to which sulphate aerosol in the troposphere and stratosphere is a product of pollutant SO_2 rather than part of the natural sulphur cycle should be determined. One attack on the problem might be by detailed work on the chemistry of sulphur-particulate pollution analogous to that which has thrown light on the photochemical smog process (see Sections 2.2, 2.4).

The deposition of sulphate ion in rain is an associated problem with a long-term biological interest. Careful and detailed long-term investigation of precipitation chemistry as part of an ecological program is indicated. Forest and lake ecology in Northeastern U.S.A. and Scandinavia are sensitive areas (see Section 7).

Regional and sub-continental air pollution models are of interest in this connection (see Section 4).

9.1.4 Nitrogen Oxides—Transport and Life Cycle

There is possible interest in the long-term biological effects of nitrate rain-out. The problems are similar to those for SO_2. A quantitative budget of pollutant nitrogen in the Los Angeles area would be interesting (see Sections 2.5, 7).

9.1.5 H_2O Concentration

Long-term effects of water emission from surface sources seem likely to be very localized but two aspects of the emissions from aircraft cause concern—a possible increase in cloud caused by present generation machines and a probable significant increase in stratospheric H_2O concentration if supersonic transport becomes general. Careful organized observation would help with the first problem. A minimal monitoring program for stratospheric water vapor could be initiated now. The computational investigation of radiative flux divergence is in its least satisfactory state near the tropopause (see Section 2.7).

9.1.6 Carbon Monoxide—Concentration, Transport and Life Cycle

Very recent theoretical work has opened a new aspect of this problem. No serious geophysical or biological long-term effect of CO emission has been identified (see Section 2.3).

9.2 Long-term Climatic Effects

Empirical correlation of observed climatic trends with observed pollution trends does not establish causal connection (see Section 3.1). We must seek a physical theory, by way of climatic models. The simpler models will probably always be inadequate, but are suggestive. Current examples have weaknesses in the treatment of meridional energy transport and the relation of albedo to temperature (see Section 3.2). The more complex current models are deficient in their treatment of cloud and its reaction on the radiation field and albedo. Dynamically, they may be more elaborate than is necessary for the long-term climatic application. Proper treatment of the effects of aerosol will always be a major difficulty (see Sections 3.3, 3.4).

The more obvious aspects of climate are effectively monitored by weather services, but the records should be examined for regional changes in areas subject to changing pollution levels.

One aspect of climatic change which could be examined by detailed modeling is the possibility of global effects following a regional change in some dynamically sensitive area—e.g., a change in albedo or radiative flux divergence over the land areas of eastern North America (see Section 3.3).

9.3 Other Long-term Geophysical Effects

Changes in the stratosphere and mesosphere are an aspect of climatic change, but they may also involve changes in the lower ionosphere with consequent changes in radio-propagation. The effect of changes of CO_2 content on the temperature of the mesosphere should be further investigated in the light of possible changes in chemical reaction rates. It seems unlikely that surface-emitted NO could reach the mesosphere, where it would be very reactive. Changes of H_2O content would affect the rates of some photochemical processes (see Sections 2.1, 2.3, 2.7).

9.4 Solar Radiation and Radiative Transfer (Other than as a Facet of Climatic Models)

The reduction of solar radiation by polluted atmospheres has not been shown to have any major biological effects at its present level. It is severe at times and might

have subtle psychological and socio-economic effects, so deserves examination in its own right. It can also serve, suitably interpreted, as a monitor of aerosol content and to provide indirect estimates of atmospheric albedo and absorption. As an instrument for monitoring slow global changes remote, preferably high-altitude, stations are called for. As a minimum they should record direct and diffuse solar radiation in the U.V., visible, and I.R. They must pay careful attention to standardization. Diffuse radiation in the I.R. is a sensitive indicator of aerosol loading (see Section 5.2).

The network of simple "turbidity" measurements established by NAPCA monitors the geographical distribution of aerosol pollution, and should indicate statistically any secular changes. It should be expanded geographically, and maintained. Calibration standards must not be relaxed (see Section 5.2).

Turbidity measurements give a relative measure of total aerosol loading. They should be supplemented in a few locations remote from pollution sources by observations of the height distribution of aerosol using stable or well calibrated methods. Laser ('Lidar') methods appear to offer a good approach. The objective is stable long-term monitoring of the aerosol loading in the upper troposphere and lower stratosphere (see Sections 5.2, 8).

9.5 Nucleation and Precipitation

9.5.1 Nuclei Concentrations

There is insufficient information on the concentrations of ice and condensation nuclei as related to various anthropogenic sources. There is a particular need for more detailed data on the removal processes which operate in the atmosphere. It is vital to determine the areal and quantitative extent of the nuclei changes now taking place in the atmosphere and to isolate the possible long-term buildups in concentration from the local variability. Two scales of nuclei observations should be considered:

a. Global scale—A fixed network of up to 10 stations should be established to measure condensation and ice nuclei on a routine basis. A portion of these should be located in relatively clean air, i.e., Hawaii and non-populated sections of the West Coast. A portion of the network should also be located in the Northeastern United States where local nuclei levels may be increasing regardless of the trend on a global basis.

b. Local scale—In addition to the fixed network of nuclei stations, it would be useful to have mobile units which could explore the effects of individual source areas, determine their downwind extent, and obtain useful information on the effectiveness of the removal processes.

(see Sections 2.2, 6.2, 6.3, 6.4).

9.5.2 Observational Studies

Primary evidence of inadvertent effects has come from statistical studies of climatological data. Several areas of possible effect have been identified in this

manner. Further progress in these cases will come from the development of a better physical understanding of the causes of the anomalies. This can come about through short, intensive field studies of nuclei, heat, and moisture fields in the area of interest, together with modeling studies.

A detailed study of the environment in the areas of possible inadvertent effect, e.g., La Porte or Seattle, should be made (see Section 6.5.1).

The variability in cloud droplet concentrations (particularly in summer cumulus) should be explored for various areas of the country. It would be particularly important to obtain more concentration data in the east and northeast United States where numerous particulate sources may combine to influence large sections. Coalescence processes in summer cumulus should be studied, requiring concurrent radar, cloud nuclei, and cloud drop measurements (see Sections 6.5.1, 6.5.2).

Additional data are needed on free iodine concentrations in the atmosphere in order to determine the importance of the automobile exhaust-iodine reaction (see Section 6.4).

9.5.3 Model Studies

Many of the processes involved in the inadvertent nucleation effects can be modeled with computer studies. This includes the coalescence process, plumes from isolated sources, frictional and thermal "heat island" effects, and dynamic seeding effects of ice nuclei. A very considerable effort is already being expended but there is room for additional modeling work, particularly directed toward simple model frameworks which can be used for parametric sensitivity studies (see Section 6.5).

9.5.4 Statistical Studies

Much has already been accomplished as a result of statistical studies in calling attention to the problem of inadvertent modification. Additional studies in other areas and in greater depth in already-identified locations are warranted (see Section 6.5).

9.5.5 Laboratory Studies

It seems inevitable that many of the inadvertent modification problems will have to be resolved using in situ atmospheric measurements. Supporting laboratory studies would be valuable, however. These should include additional work on automobile exhausts aimed at determining required iodine concentrations, studies of nucleating properties of various materials and pollution processes, and the general problem of the changes in nucleation properties due to agglomeration, radiation, etc., which occur in the atmosphere after release from the source (see Section 6).

9.6 Applications of Artificial Earth Satellites

Weather satellite systems introduce the possibility of monitoring albedo. It seems likely that the precision attainable by present systems is insufficient; this should be studied and improved when possible. Projected systems for global temperature observation may bring an opportunity for CO_2 and aerosol monitoring (see Sections 3, 8.2).

In general, satellite methods do not seem obviously suited to investigation of long-term air pollution problems—the enormous data-gathering capacity does not appear to be particularly advantageous. Cost benefit analysis should be made of all proposed satellite measurements, but the possibilities of combining with other monitoring on weather and earth resources satellites should be kept in mind by active feasibility studies (see Section 8.2).

10.0 PROJECTS FOR EARLY ATTENTION

Priorities must be assessed by a compromise between importance, likelihood of early success, and magnitude and expense of the effort involved in relation to the available total. I suggest that first attention should be given to the monitoring projects which have begun, to ensure continuance, to allow a modest expansion, and to improve methods. These projects are CO_2 measurement at a remote station, solar radiation measurement at a remote station, and the widespread measurement of atmospheric turbidity. The addition of measurements of all types of nuclei at one or more remote stations should be considered at this stage, as should proposals for the monitoring of stratospheric and high tropospheric aerosol, perhaps by "lidar" methods.

NAPCA should also support projects for monitoring the planetary albedo, particularly by measurement from satellites, though I doubt whether the necessary precision will be attained at an early date.

Comprehensive monitoring of the chemical composition of precipitation, in conjunction with ecological studies is suggested as a first step in the study of truly long-term biological effects of pollution.

Turning from observation to the areas of understanding and prediction, there is a requirement for further development of climatic modeling. NAPCA's particular interest is in the long-term response to small but specific changes in atmospheric constitution, and concentration on this aspect calls for some shift in the priorities of groups presently engaged in this arduous and expensive work. Resources are stretched even now, and an early review of facilities and potential by all interested agencies might be useful.

Long-term effects on less than global scale are likely to cause increasing public concern. The sub-continental scale transport of pollutants might be studied by an extension of urban air pollution models or modification of weather forecasting models. No great scientific advance is involved; the political interest suggests early action.

More localized effects can properly be described as potentially long-term modifications of climate. Urban effects on precipitation patterns and solar radiation can properly be studied within a program concerned with "long-term geophysical and biological effects of pollution." The relative ease of formulating specific projects, and the increased probability of an early useful outcome should not be allowed to divert too great a proportion of support from the more difficult larger-scale and longer-term problems which will not be quickly solved.

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